



Patent
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Richard F. Lemuth
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Date: Oct. 17, 2006

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF APPEALS AND INTERFERENCES

In re the application of)	
)	
JOHN ROBERT LOCKEMEYER ET AL)	
)	
)	
Serial No. 10/606,440)	Group Art Unit: 1755
)	
Filed June 26, 2003)	Examiner: Hailey, Patricia L.
)	
A METHOD FOR IMPROVING THE)	October 17, 2006
SELECTIVITY OF A CATALYST AND)	
A PROCESS FOR THE EPOXIDATION)	
OF AN OLEFIN)	
)	

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Sir:

APPELLANT'S BRIEF

The following brief is on appeal of a final rejection of Claims 1-9 and 11-25 of the above-identified U.S. patent application. The final rejection was contained in an Office Action mailed on February 3, 2006, and a Notice of Appeal was mailed on June 29, 2006. Please charge the appropriate fees for filing this brief to Shell Oil Company Deposit Account No. 19-1800. It is respectfully requested that the Board consider the following arguments and reverse the final rejection of Claims 1-9 and 11-25 in the above-identified application. A request for a one-month extension for reply accompanies this brief.

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Real Party of Interest:

The invention of the present application is assigned to Shell Oil Company, which is the real party of interest in the present appeal.

Related Appeals and Interferences:

Appellant, and appellant's legal representative, are not aware of any appeals or interferences that directly affect or could be directly affected by or have a bearing on the Board's decision in the present appeal.

Status of the Claims:

Claims 1-38 were presented for examination. Claim 10 was canceled. Claims 26-38 were withdrawn from consideration. Applicant reserves the right to file and prosecute a divisional application related to the subject matter of Claims 26-38. Claims 1-9 and 11-25 are the subject of the present Appeal and stand rejected by the Examiner.

Status of Amendments:

All amendments were entered prior to the final rejection. No amendments were filed after the mailing of the Final Office Action.

Summary of Claimed Subject Matter:

The present invention provides a solution to the problem of improving the selectivity of certain supported high selectivity catalysts having a relatively low silver density, in particular catalysts containing silver in a quantity of at most 0.17 g per m² surface area of the support and one or more selectivity enhancing dopants (rhenium, molybdenum, and tungsten, as defined in claim 1). *See U.S. Patent Application Serial No. 10/606,440* (hereinafter "*Application*"), page 2, lines 23-33; page 3, lines 19-23; page 7, lines 28-31; page 8, lines 10-16. An improvement in selectivity was found by contacting the catalyst, or precursor of the catalyst comprising the silver in cationic form, with a feed comprising oxygen at a catalyst temperature above 250 °C for a duration of at least 0.5 hours and up to 150 hours. *See Id.* at page 2, lines 23-33; page 3, lines 19-23; page 9, line 29 – page 10, line 3; page 11, lines 7-28. Subsequently, the catalyst temperature is decreased to a value of at most 250 °C. *See Id.* at page 13, lines 1-3.

Grounds of Rejection to be Reviewed on Appeal:

Claims 1-9 and 11-25 stand rejected under 35 U.S.C. § 103(a) as being obvious over U.S. Patent No. 4,007,135 (hereinafter the '135 reference).

Argument:

Claims 1-9 and 11-25 under 35 U.S.C. § 103(a) were rejected as being unpatentable over Hayden et al (U.S. Patent No. 4,007,135). In this case, a *prima facie* basis for obviousness has not been established.

As explained in the application text (page 7, line 28 – page 8, line 13), the present invention may specifically be applicable to epoxidation catalysts having a selectivity enhancing dopant (as defined) and having a relatively low silver density. Catalysts having a higher silver density are preferably not subjected to the treatment of the present invention. *See Application*, page 4, lines 3-8. It has been found that the selectivity of a highly selective epoxidation catalyst, as defined in Claim 1, can be improved by heat-treating the catalyst in the presence of oxygen at a temperature which is typically above the catalyst's normal initial operation temperature. *See Application*, page 3, lines 19-23

The M.P.E.P. § 2143 states:

To establish a *prima facie* case of obviousness, three basic criteria must be met. First, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify the reference or to combine reference teachings. Second, there must be a reasonable expectation of success. Finally, the prior art reference (or references when combined) must teach or suggest all the claim limitations.

The teaching or suggestion to make the claimed combination and the reasonable expectation of success must both be found in the prior art, not in applicant's disclosure. *In re Vaeck*, 947 F.2d 488, 20 U.S.P.Q.2d 1438 (Fed. Cir. 1991).

A single prior art reference can render a claim obvious; however, there must be a showing of a suggestion or motivation to modify the teachings of that reference to the claimed invention. *SIBIA Neurosciences, Inc., v. Cadus Pharmaceutical Corp.*, 225 F.3d 1349, 1356 (Fed. Cir. 2000). All of the reference's teachings must be considered. *W.L. Gore & Assoc., Inc. v. Garlock, Inc.*, 721 F.2d 1540, 1550 (Fed. Cir. 1983). There can be no showing of a suggestion or motivation to modify a reference if that reference "suggests that the line of development flowing from the reference's disclosures is unlikely to be productive of the result sought by the applicant." *In re Garley*, 27 F.3d 551, 553 (Fed. Cir. 1994).

In its generic disclosures, the '135 reference teaches improved catalysts for the production of alkylene oxides containing silver and an additional promoter supported on a

heat resisting support which has a specific surface area in the range 0.04 to 10 m²/g, especially 0.2 to 0.6 m²/g, an apparent porosity of at least 20 %, and median pore diameter of 0.3 to 15 microns. *U.S. Patent No. 4,007,135* col. 1, ll. 20-41. The catalyst preferably comprises 3 to 15 % by weight silver, more preferably 6 to 12 % by weight silver. *Id.* at col. 3, ll. 27-28. There are many possible promoters disclosed in the '135 reference. For example, possible promoters include: copper, gold, zinc, cadmium, mercury, niobium, tantalum, molybdenum, tungsten, vanadium, or preferably chromium, calcium, magnesium, strontium and/or, more preferably barium. *Id.* at col. 1, ll. 35-39. Additional promoters include: alkali metals, especially sodium, potassium or rubidium. *Id.* at col. 3, ll. 13-17. Also, the '135 reference discloses many different properties for the support such as surface area, apparent porosity, median pore diameter, pore size distribution, pore volume, ratio of median pore diameter to average equivalent diameter of the silver particles. *Id.* at col 1, ll. 25-35; col. 3, ll. 28-50. Further, the '135 reference generically discloses decomposing the silver salt (compound) by heating to a temperature of 200-400 °C. *Id.* at col. 5, ll. 17-20. There is no generic disclosure with respect to heating atmospheres or heating times.

When considering the '135 reference as a whole, the working examples should also be considered in addition to the generic disclosures. With respect to decomposition conditions, Examples 4 and 10 of the '135 reference show the use of lower decomposition temperatures as well as higher decomposition temperatures to reduce the silver salt (compound) without indicating a preference. Example 4 discloses reducing the silver salt in an atmosphere comprising hydrogen at a maximum temperature of 220 °C. *U.S. Patent No. 4,007,135*, col. 9, ll. 15-20. Example 10 discloses reducing the silver salt by heating in a forced draught oven for 4 hours while the temperature was raised from 100 to 300 °C at a rate of 0.8 °C/min. *Id.* at col. 12, ll. 28-32.

Further, a comparison of Example 7 to Example 27 shows that decomposing silver using higher temperatures with catalysts having silver densities greater than claimed in the present invention can lead to a decline in catalyst selectivity. In Example 7, the silver impregnated supports are heated in a forced draught oven for 4 hours while the temperature is raised from 100 to 280 °C at a rate of 0.8 °C/min. The catalysts in Example 7 contained 8 % by weight silver and were prepared using a support having a surface area of 0.17 m²/g. *Id.* at col. 9, l. 65 – col. 10, l. 2. The silver density can be calculated to be 0.47 grams of silver

per m² surface area of the support which is greater than claimed in the present invention. None of the catalysts in Example 7 contained rhenium, molybdenum or tungsten as catalytic components. In Example 27, the silver impregnated supports are heated in a forced draught oven for 4 hours while the temperature is raised from 100 to 300 °C at a rate of 0.8 °C/min. *Id.* at col. 21, ll. 36-41. The disclosures in Example 7 and 27 appear to be the same except for the maximum silver decomposition temperatures (280 °C vs. 300 °C) and the preparation of the silver solutions. The silver solution in Example 7 contained ethanolamine while it is not mentioned in Example 27. Catalysts J, P, R, and T of Example 7 disclose the same catalytic components as Catalysts 18', 29', 20', and 21', respectively, of Example 27. In comparing the selectivity data disclosed for these catalysts, one skilled in the art would find at lower conversion levels the effect of heating to higher decomposition temperatures is ambiguous at best, while at the higher conversion level, i.e., 40 % oxygen conversion, the higher decomposition temperature significantly reduces the selectivity of the catalyst. This is clearly demonstrated in Table I below. In particular, the selectivity was over 5 percent lower for Catalysts 18' and 29' of Example 27 which were heated to 300 °C as compared to Catalysts J and P of Example 7 which were heated to 280 °C.

Table I

Catalyst (from Example)	Additive	Atoms of additive per 1000 atoms Ag	Maximum decomposition temperature, °C	Selectivity at 40 % oxygen conversion, %-mole	Difference, %-mole ¹⁾
J (7)	None	None	280	78	
18' (27)	None	None	300	72.9	- 5.1
P (7)	Na	69	280	90.6	
29' (27)	Na	69	300	85.4	- 5.2
R (7)	Na	692	280	90.1	
20' (27)	Na	692	300	87.1	- 3
T (7)	K	70	280	85.8	
21' (27)	K	70	300	85.2	- 0.6
¹⁾ Selectivity obtained after decomposition at 300 °C maximum minus selectivity obtained after decomposition at 280 °C maximum					

The '135 reference, when considering the teachings as a whole, provides generically no preference to use higher decomposition temperatures over lower decomposition temperatures or lower decomposition temperatures over higher decomposition temperatures when drying the catalyst and the working examples suggest lower decomposition

temperatures are better for catalyst selectivity, see Table I above. The '135 reference would suggest to one skilled in the art to use lower decomposition temperatures (i.e., below 250 °C), such as 220 °C as described in Example 4.

The Examiner states: "The fact that Hayden et al. [(the '135 reference)] do not 'distinguish between high or low temperatures for decomposing silver to silver metal, high or low silver densities of the catalysts, or the various promoters utilized with the catalysts' does not preclude the reference from reading upon the instantly claimed invention." *Advisory Action, mailed August 17, 2006*, continuation sheet, 1st paragraph. However, to establish obviousness, the examiner must show there is a suggestion or motivation to modify the teachings of a prior art reference. The examiner "must do more than identify the elements in the prior art." *In re Huston*, 308 F.3d 1267, 1280 (Fed. Cir. 2002). A showing of a suggestion or motivation is required since most inventions are combinations of old elements. *Environmental Designs, Ltd. v. Union Oil Co. of California*, 713 F.2d 693, 698 (Fed. Cir. 1983). The Examiner identifying the elements of the claimed invention in the prior art reference does not provide sufficient suggestion or motivation to modify its teaching.

Further, there can be no showing of a suggestion or motivation to modify the '135 reference since the '135 reference suggests that using higher temperatures to decompose silver is unlikely to be productive of the result sought, i.e., an improvement in selectivity, and that using lower temperatures to decompose silver is more likely to result in an improvement in selectivity. The '135 reference does not teach or suggest modifying the '135 reference to improve catalyst selectivity by utilizing the combination of a catalyst having a relatively low silver density in addition to selectivity enhancing dopants and heating of the silver-containing catalyst, or precursor of the catalyst containing silver in cationic form, in the presence of oxygen at a certain temperature for a certain period of time as specified in claim 1. The '135 reference does not teach or suggest any relationship between the amount of silver and the surface area of the support, i.e., silver density. Additionally, the '135 reference does not teach or suggest any relationship between particular values for the silver density, promoters and heating conditions.

Therefore, there is no suggestion or motivation to modify the '135 reference to arrive at the method of the present invention.

In order to establish a *prima facie* case of obviousness, there must also be a reasonable expectation of success for the modification of the prior art. M.P.E.P. §§ 2143, 2143.02. The Examiner asserts that it would have been obvious to one skilled in the art at the time the invention was made to reasonably expect that the method of the '135 reference would result in improved catalyst selectivity, in view of the strong similarities between the method steps, conditions, and catalyst components of the '135 reference and the claimed invention. *Office Action, mailed April 3, 2006*, page 4, 2nd paragraph; page 5, 5th paragraph – page 6, 2nd paragraph.

With respect to the silver decomposition method, as discussed above, the '135 reference provides generically no preference to use higher decomposition temperatures over lower decomposition temperatures or lower decomposition temperatures over higher decomposition temperatures when drying the catalyst and the working examples suggest lower decomposition temperatures are better for catalyst selectivity. The working Examples 7 and 27 of the '135 reference (See Table I, above) would suggest to one skilled in the art to use lower decomposition temperatures (i.e., below 250 °C), such as 220 °C as described in Example 4. One skilled in the art would not reasonably expect the silver decomposition method of the '135 reference would result in an additional improvement in catalyst selectivity from using higher rather than lower decomposition temperatures.

Additionally, the Examiner is clearly applying an improper “obvious to try” rationale in support of the obviousness rejection. M.P.E.P. § 2145(X)(B). Obvious to try has been defined in terms of what “would have been to vary all parameters or try each of numerous possible choices until one possibly arrived at a successful result, where the prior art gave either no indication of which parameters were critical or no direction as to which of many possible choices is likely to be successful....” *Id.*

Care must be taken not “to fall victim to the insidious effect of a hindsight syndrome wherein that which only the inventor taught is used against its teacher.” *W.L. Gore*, 721 F.2d at 1553. Bits and pieces of the invention as currently claimed may be pointed to in the prior art, but only if one is armed with hindsight knowledge and does not consider all the teachings in the prior art reference. The '135 reference does no more than suggest experimenting with many possible combinations of parameters. ‘Obvious to try’ is not the standard for obviousness.

The '135 reference discloses many different properties with respect to the catalyst and the support, as discussed hereinbefore. In particular, many possible promoters are disclosed as well as many different properties for the support, for example surface area, apparent porosity, median pore diameter, pore size distribution pore volume, ratio of median pore diameter to average equivalent diameter of the silver particles. The generic teachings of the '135 reference do not provide an indication of which parameters are critical or which direction as to the many possible choices is likely to be successful. As discussed above, the '135 reference does not teach or suggest any relationship between the amount of silver and the surface area of the support, i.e., silver density. Further, the '135 reference does not teach or suggest any relationship between particular values for the silver density, promoters and heating conditions to indicate which of the many possible combinations of parameters would be successful.

Therefore, it would not have been obvious to one skilled in the art when considering the '135 reference as a whole to reasonably expect that a specific treatment for a certain group of epoxidation catalysts, as defined in claim 1 by silver density and selected selectivity enhancing dopants, would result in a distinct advantage in selectivity.

Also, the '135 reference does not teach or suggest all the claim limitations. In particular, as discussed hereinbefore, the '135 reference does not teach or suggest the combination of silver density, selectivity enhancing dopants and heating of the silver-containing catalyst, or precursor of the catalyst containing silver in cationic form, in the presence of oxygen at a certain temperature for a certain period of time, as specified in the claims.

In view of these arguments, Applicants respectfully submit that a *prima facie* case of obviousness has not been established for Claims 1-9 and 11-25. Therefore, these claims are unobvious and patentable over the '135 reference.

In conclusion, for the reasons set forth above, the Applicants request that the Board overturn the Examiner's rejection.

Respectfully submitted,

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Claims Appendix:

1. (Previously Presented) A method for improving the selectivity of a supported highly selective epoxidation catalyst comprising silver in a quantity of at most 0.17 g per m² surface area of the support and further comprising one or more selectivity enhancing dopants selected from rhenium, molybdenum, and tungsten, which method comprises
 - contacting the catalyst, or a precursor of the catalyst comprising the silver in cationic form, with a feed comprising oxygen at a catalyst temperature above 250 °C for a duration of at least 0.5 hours and up to 150 hours, and
 - subsequently decreasing the catalyst temperature to a value of at most 250 °C.
2. (Original) A method as claimed in claim 1, wherein the catalyst, or the precursor of the catalyst, comprises an α -alumina support having a surface area of at least 1 m²/g, and a pore size distribution such that pores with diameters in the range of from 0.2 to 10 μ m represent at least 70 % of the total pore volume and such pores together provide a pore volume of at least 0.25 ml/g, relative to the weight of the support.
3. (Previously Presented) A method as claimed in claim 1, wherein the catalyst, or the precursor of the catalyst, comprises, in addition to silver, a Group IA metal.
4. (Previously Presented) A method as claimed in claim 3, wherein the selectivity enhancing dopant is rhenium; and wherein the catalyst, or the precursor of the catalyst, further comprises a further metal or compound thereof selected from the group of Group IA metals, Group IIA metals, molybdenum, tungsten, chromium, titanium, hafnium, zirconium, vanadium, thallium, thorium, tantalum, niobium, gallium and germanium and mixtures thereof, and optionally a rhenium co-promoter which may be selected from one or more of sulfur, phosphorus, boron, and compounds thereof, on the support material.
5. (Original) A method as claimed in claim 1, wherein the catalyst, or the precursor of the catalyst, comprises silver in a quantity of from 50 to 250 g/kg, on the total catalyst, and the support has a surface area of from 1 to 5 m²/g.
6. (Original) A method as claimed in claim 1, wherein the feed comprises oxygen in a quantity of from 1 to 30 %v, relative to the total feed.

7. (Original) A method as claimed in claim 6, wherein the feed comprises oxygen in a quantity of from 2 to 25 %v, relative to the total feed.
8. (Original) A method as claimed in claim 1, wherein the catalyst, or the precursor of the catalyst, is contacted with a feed comprising oxygen at a temperature in the range of from 250 to 320 °C.
9. (Original) A method as claimed in claim 8, wherein the temperature in the range of from 255 to 300 °C.
10. (Canceled)
11. (Original) A method as claimed in claim 10, wherein the duration is in the range of from 1 to 50 hours.
12. (Original) A method as claimed in claim 11, wherein the duration is in the range of from 2 to 40 hours.
13. (Original) A method as claimed in claim 1, wherein the feed comprises an olefin, in addition to oxygen.
14. (Original) A method as claimed in claim 13, wherein the feed comprises the olefin in a concentration of from 0.5 to 70 mole-%; oxygen in a concentration of from 1 to 15 mole-%; and, in addition, a saturated hydrocarbon, if any, in a concentration of from 0 to 80 mole-%, relative to the total feed.
15. (Original) A method as claimed in claim 14, wherein the feed comprises the olefin in a concentration of from 1 to 60 mole-%; oxygen in a concentration of from 2 to 10 mole-%; and a saturated hydrocarbon in a concentration of from 30 to 75 mole-%, relative to the total feed.
16. (Original) A method as claimed in claim 1, wherein the quantity of silver relative to the surface area of the support is in the range of from 0.01 to 0.15 g/m².
17. (Original) A method as claimed in claim 16, wherein the quantity of silver relative to the surface area of the support is in the range of from 0.02 to 0.12 g/m².

18. (Original) A method as claimed in claim 16, wherein the catalyst, or the precursor of the catalyst, comprises silver in a quantity of from 50 to 250 g/kg, on the total catalyst, and the catalyst comprises an α -alumina support having a surface area of from 1 to 5 m²/g, and a pore size distribution such that pores with diameters in the range of from 0.2 to 10 μ m represent at least 70 % of the total pore volume and such pores together provide a pore volume of at least 0.25 ml/g, relative to the weight of the support.

19. (Previously Presented) A method as claimed in claim 16, wherein the catalyst, or the precursor of the catalyst, comprises, in addition to silver, a Group IA metal.

20. (Previously Presented) A method as claimed in claim 19, wherein the selectivity enhancing dopant is rhenium; and wherein the catalyst, or the precursor of the catalyst, further comprises a further metal or compound thereof selected from the group of Group IA metals, Group IIA metals, molybdenum, tungsten, chromium, titanium, hafnium, zirconium, vanadium, thallium, thorium, tantalum, niobium, gallium and germanium and mixtures thereof, and optionally a rhenium co-promoter which may be selected from one or more of sulfur, phosphorus, boron, and compounds thereof, on the support material.

21. (Original) A method as claimed in claim 16, wherein the catalyst, or the precursor of the catalyst, is contacted with a feed comprising oxygen at a temperature in the range of from 250 to 320 °C.

22. (Original) A method as claimed in claim 21, wherein the catalyst, or the precursor of the catalyst, is contacted with a feed comprising oxygen at a temperature in the range of from 255 to 300 °C.

23. (Original) A method as claimed in claim 16, wherein the duration is in the range of from 1 to 50 hours.

24. (Original) A method as claimed in claim 23, wherein the duration is in the range of from 2 to 40 hours.

25. (Original) A method as claimed in claim 24, wherein the feed comprises the olefin in a concentration of from 0.5 to 70 mole-%; oxygen in a concentration of from 1 to 15 mole-%; and, in addition, a saturated hydrocarbon, if any, in a concentration of from 0 to 80 mole-%, relative to the total feed.

26. (Withdrawn) A process for the epoxidation of an olefin, which process comprises the steps of

- contacting a supported highly selective epoxidation catalyst comprising silver in a quantity of at most 0.17 g per m² surface area of the support, or a precursor of the catalyst comprising the silver in cationic form, with a feed comprising oxygen at a catalyst temperature above 250 °C for a duration of up to 150 hours, and
- subsequently decreasing the catalyst temperature to a value of at most 250 °C and contacting the catalyst with the feed comprising the olefin and oxygen.

27. (Withdrawn) A method as claimed in claim 26, wherein the quantity of silver relative to the surface area of the support is in the range of from 0.01 to 0.15 g/m².

28. (Withdrawn) A method as claimed in claim 27, wherein the quantity of silver relative to the surface area of the support is in the range of from 0.02 to 0.12 g/m².

29. (Withdrawn) A method as claimed in claim 27, wherein the catalyst, or the precursor of the catalyst, comprises silver in a quantity of from 50 to 250 g/kg, on the total catalyst, and the catalyst, or the precursor of the catalyst, comprises an α -alumina support having a surface area of from 1 to 5 m²/g, and a pore size distribution such that pores with diameters in the range of from 0.2 to 10 μ m represent at least 70 % of the total pore volume and such pores together provide a pore volume of at least 0.25 ml/g, relative to the weight of the support.

30. (Withdrawn) A method as claimed in claim 29, wherein the catalyst, or the precursor of the catalyst, comprises, in addition to silver, rhenium or compound thereof, a further metal or compound thereof selected from the group of Group IA metals, Group IIA metals, molybdenum, tungsten, chromium, titanium, hafnium, zirconium, vanadium, thallium, thorium, tantalum, niobium, gallium and germanium and mixtures thereof, and optionally a rhenium co-promoter which may be selected from one or more of sulfur, phosphorus, boron, and compounds thereof, on the support material.

31. (Withdrawn) A method as claimed in claim 26, wherein in the step in which the catalyst, or the precursor of the catalyst, is contacted at a temperature above 250 °C with a feed comprising oxygen, the temperature is selected in the range of from 255 to 320 °C.

32. (Withdrawn) A method as claimed in claim 26, wherein the duration is in the range of from 1 to 50 hours.
33. (Withdrawn) A method as claimed in claim 26, wherein in the step in which the catalyst, or the precursor of the catalyst, is contacted at a temperature above 250 °C with a feed comprising oxygen, the feed comprises the olefin in a concentration of from 0.5 to 70 mole-%; oxygen in a concentration of from 1 to 15 mole-%; and a saturated hydrocarbon, if any, in a concentration of from 0 to 80 mole-%, relative to the total feed.
34. (Withdrawn) A process as claimed in claim 26, wherein the olefin is ethylene.
35. (Withdrawn) A process as claimed in claim 26, wherein the feed comprising the olefin and oxygen comprises in addition, as a reaction modifier, an organic chloride and optionally a nitrate- or nitrite-forming compound.
36. (Withdrawn) A process as claimed in claim 26, wherein the catalyst temperature is decreased to a value in the range of from 180 to 250 °C.
37. (Withdrawn) A process as claimed in claim 36, wherein the catalyst temperature is decreased to a value in the range of from 200 to 245 °C.
38. (Withdrawn) A process for producing a 1,2-diol, 1,2-diol ether, or an alkanolamine, comprising converting an olefin oxide into the 1,2-diol, the 1,2-diol ether, or the alkanolamine, wherein the olefin oxide has been obtained by a process for the epoxidation of an olefin according to claim 26.